

## PATENT COOPERATION TREATY

PCT

## NOTIFICATION OF ELECTION

(PCT Rule 61.2)

From the INTERNATIONAL BUREAU

To:

Assistant Commissioner for Patents  
 United States Patent and Trademark  
 Office  
 Box PCT  
 Washington, D.C.20231  
 ÉTATS-UNIS D'AMÉRIQUE

in its capacity as elected Office

<b>Date of mailing</b> (day/month/year) 13 September 1999 (13.09.99)	
<b>International application No.</b> PCT/GB98/03636	<b>Applicant's or agent's file reference</b> P/3952.WO
<b>International filing date</b> (day/month/year) 07 December 1998 (07.12.98)	<b>Priority date</b> (day/month/year) 05 December 1997 (05.12.97)
<b>Applicant</b> CHOY, Kwang-Leong et al	

1. The designated Office is hereby notified of its election made:



in the demand filed with the International Preliminary Examining Authority on:

23 June 1999 (23.06.99)



in a notice effecting later election filed with the International Bureau on:

2. The election ☒ was

was not

made before the expiration of 19 months from the priority date or, where Rule 32 applies, within the time limit under Rule 32.2(b).

The International Bureau of WIPO  
 34, chemin des Colombettes  
 1211 Geneva 20, Switzerland

Facsimile No.: (41-22) 740.14.35

Authorized officer

C. Carrié

Telephone No.: (41-22) 338.83.38

# PCT

## INTERNATIONAL SEARCH REPORT

(PCT Article 18 and Rules 43 and 44)

Applicant's or agent's file reference <b>P/3952.WO</b>	<b>FOR FURTHER ACTION</b> see Notification of Transmittal of International Search Report (Form PCT/ISA/220) as well as, where applicable, item 5 below.	
International application No. <b>PCT/GB 98/ 03636</b>	International filing date (day/month/year) <b>07/12/1998</b>	(Earliest) Priority Date (day/month/year) <b>05/12/1997</b>
Applicant <b>IMPERIAL COLLEGE OF SCIENCE, TECHNOLOGY AND MEDICI</b>		

This International Search Report has been prepared by this International Searching Authority and is transmitted to the applicant according to Article 18. A copy is being transmitted to the International Bureau.

This International Search Report consists of a total of 4 sheets.

☒ It is also accompanied by a copy of each prior art document cited in this report.

### 1. Basis of the report

- a. With regard to the **language**, the international search was carried out on the basis of the international application in the language in which it was filed, unless otherwise indicated under this item.

☐ the international search was carried out on the basis of a translation of the international application furnished to this Authority (Rule 23.1(b)).

- b. With regard to any **nucleotide and/or amino acid sequence** disclosed in the international application, the international search was carried out on the basis of the sequence listing :

☐ contained in the international application in written form.

☐ filed together with the international application in computer readable form.

☐ furnished subsequently to this Authority in written form.

☐ furnished subsequently to this Authority in computer readable form.

☐ the statement that the subsequently furnished written sequence listing does not go beyond the disclosure in the international application as filed has been furnished.

☐ the statement that the information recorded in computer readable form is identical to the written sequence listing has been furnished

2. ☐ **Certain claims were found unsearchable** (See Box I).

3. ☐ **Unity of invention is lacking** (see Box II).

4. With regard to the **title**,

☒ the text is approved as submitted by the applicant.

☐ the text has been established by this Authority to read as follows:

5. With regard to the **abstract**,

☐ the text is approved as submitted by the applicant.

☒ the text has been established, according to Rule 38.2(b), by this Authority as it appears in Box III. The applicant may, within one month from the date of mailing of this international search report, submit comments to this Authority.

6. The figure of the **drawings** to be published with the abstract is Figure No.

☒ as suggested by the applicant.

☐ because the applicant failed to suggest a figure.

☐ because this figure better characterizes the invention.

1

☐ None of the figures.

# INTERNATIONAL SEARCH REPORT

International application No.

PCT/GB 98/ 03636

Box III TEXT OF THE ABSTRACT (Continuation of item 5 of the first sheet)

The abstract has to be changed as follows:  
Line 5, after "substrate" insert "(50)";  
line 6, after "outlet" insert "(60)";  
line 7, after "substrate" insert "(50)".

# INTERNATIONAL SEARCH REPORT

International Application No  
PCT/GB 98/03636

## A. CLASSIFICATION OF SUBJECT MATTER

IPC 6 C23C16/44

According to International Patent Classification (IPC) or to both national classification and IPC

## B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)  
IPC 6 C23C

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

## C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category °	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	PATENT ABSTRACTS OF JAPAN vol. 005, no. 054 (C-050), 15 April 1981 & JP 56 005337 A (HITACHI LTD; OTHERS: 01), 20 January 1981	1,2,6,7, 9-12,16
Y	see abstract	3,4,14, 15 8
A	---	
X	PATENT ABSTRACTS OF JAPAN vol. 006, no. 143 (C-117), 3 August 1982 & JP 57 067038 A (NIPPON TELEGR & TELEPH CORP), 23 April 1982 see abstract	1,2,6,9, 16
	---	
	-/--	



Further documents are listed in the continuation of box C.



Patent family members are listed in annex.

### ° Special categories of cited documents :

- "A" document defining the general state of the art which is not considered to be of particular relevance
- "E" earlier document but published on or after the international filing date
- "L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)
- "O" document referring to an oral disclosure, use, exhibition or other means
- "P" document published prior to the international filing date but later than the priority date claimed

"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention

"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone

"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art.

"&" document member of the same patent family

Date of the actual completion of the international search

26 May 1999

Date of mailing of the international search report

02/06/1999

Name and mailing address of the ISA

European Patent Office, P.B. 5818 Patentlaan 2  
NL - 2280 HV Rijswijk  
Tel. (+31-70) 340-2040, Tx. 31 651 epo nl,  
Fax: (+31-70) 340-3016

Authorized officer

Ekhult, H

# INTERNATIONAL SEARCH REPORT

International Application No

PCT/GB 98/03636

## C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

Category °	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
Y	PATENT ABSTRACTS OF JAPAN vol. 098, no. 001, 30 January 1998 & JP 09 235131 A (SHOWA ELECTRIC WIRE & CABLE CO LTD; NIPPON TELEGR & AMP; TELEPH CORP), 9 September 1997 see abstract ---	3,4
Y	PATENT ABSTRACTS OF JAPAN vol. 014, no. 227 (C-0718), 15 May 1990 & JP 02 055241 A (HITACHI CABLE LTD), 23 February 1990 see abstract ---	14,15
A	WO 97 21848 A (BAI WEI ;CHOY KWANG LEONG (GB); IMPERIAL COLLEGE (GB)) 19 June 1997 cited in the application ---	5
A	PATENT ABSTRACTS OF JAPAN vol. 013, no. 262 (C-608), 16 June 1989 & JP 01 065040 A (SUMITOMO ELECTRIC IND LTD), 10 March 1989 see abstract -----	13

# INTERNATIONAL SEARCH REPORT

Information on patent family members

International Application No

PCT/GB 98/03636

Patent document cited in search report	Publication date	Patent family member(s)	Publication date
W0 9721848 A	19-06-1997	GB 2308132 A	18-06-1997
		AU 1182997 A	03-07-1997
		CA 2240625 A	19-06-1997
		EP 0870075 A	14-10-1998
<hr/>			

# PCT

## INTERNATIONAL SEARCH REPORT

(PCT Article 18 and Rules 43 and 44)

Applicant's or agent's file reference <b>P/3952.WO</b>	<b>FOR FURTHER ACTION</b> see Notification of Transmittal of International Search Report (Form PCT/ISA/220) as well as, where applicable, item 5 below.	
International application No. <b>PCT/GB 98/ 03636</b>	International filing date (day/month/year) <b>07/12/1998</b>	(Earliest) Priority Date (day/month/year) <b>05/12/1997</b>
Applicant <b>IMPERIAL COLLEGE OF SCIENCE, TECHNOLOGY AND MEDICI</b>		

This International Search Report has been prepared by this International Searching Authority and is transmitted to the applicant according to Article 18. A copy is being transmitted to the International Bureau.

This International Search Report consists of a total of 4 sheets.



It is also accompanied by a copy of each prior art document cited in this report.

### 1. Basis of the report

- a. With regard to the **language**, the international search was carried out on the basis of the international application in the language in which it was filed, unless otherwise indicated under this item.



the international search was carried out on the basis of a translation of the international application furnished to this Authority (Rule 23.1(b)).

- b. With regard to any **nucleotide and/or amino acid sequence** disclosed in the international application, the international search was carried out on the basis of the sequence listing :



contained in the international application in written form.



filed together with the international application in computer readable form.



furnished subsequently to this Authority in written form.



furnished subsequently to this Authority in computer readable form.



the statement that the subsequently furnished written sequence listing does not go beyond the disclosure in the international application as filed has been furnished.



the statement that the information recorded in computer readable form is identical to the written sequence listing has been furnished

2. ☐ **Certain claims were found unsearchable** (See Box I).

3. ☐ **Unity of invention is lacking** (see Box II).

### 4. With regard to the **title**,



the text is approved as submitted by the applicant.



the text has been established by this Authority to read as follows:

### 5. With regard to the **abstract**,



the text is approved as submitted by the applicant.



the text has been established, according to Rule 38.2(b), by this Authority as it appears in Box III. The applicant may, within one month from the date of mailing of this international search report, submit comments to this Authority.

6. The figure of the **drawings** to be published with the abstract is Figure No.



as suggested by the applicant.



because the applicant failed to suggest a figure.



because this figure better characterizes the invention.

1



None of the figures.

# INTERNATIONAL SEARCH REPORT

International application No.

PCT/GB 98/ 03636

Box III TEXT OF THE ABSTRACT (Continuation of item 5 of the first sheet)

The abstract has to be changed as follows:  
Line 5, after "substrate" insert "(50)";  
line 6, after "outlet" insert "(60)";  
line 7, after "substrate" insert "(50)".



## INTERNATIONAL SEARCH REPORT

International Application No

PCT/GB 98/03636

## A. CLASSIFICATION OF SUBJECT MATTER

IPC 6 C23C16/44

According to International Patent Classification (IPC) or to both national classification and IPC

## B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

IPC 6 C23C

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

## C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	PATENT ABSTRACTS OF JAPAN vol. 005, no. 054 (C-050), 15 April 1981 & JP 56 005337 A (HITACHI LTD; OTHERS: 01), 20 January 1981 see abstract	1, 2, 6, 7, 9-12, 16
Y		3, 4, 14, 15
A		8
X	PATENT ABSTRACTS OF JAPAN vol. 006, no. 143 (C-117), 3 August 1982 & JP 57 067038 A (NIPPON TELEGR & TELEPH CORP), 23 April 1982 see abstract	1, 2, 6, 9, 16



Further documents are listed in the continuation of box C.



Patent family members are listed in annex.

## \* Special categories of cited documents:

"A" document defining the general state of the art which is not considered to be of particular relevance

"E" earlier document but published on or after the international filing date

"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)

"O" document referring to an oral disclosure, use, exhibition or other means

"P" document published prior to the international filing date but later than the priority date claimed

"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention

"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone

"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art.

"&amp;" document member of the same patent family

Date of the actual completion of the international search

26 May 1999

Date of mailing of the international search report

02/06/1999

Name and mailing address of the ISA

European Patent Office, P.B. 5818 Patentlaan 2  
NL - 2280 HV Rijswijk  
Tel. (+31-70) 340-2040, Tx. 31 651 epo nl,  
Fax: (+31-70) 340-3016

Authorized officer:

Ekhult, H

## INTERNATIONAL SEARCH REPORT

International Application No

PCT/GB 98/03636

## C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
Y	PATENT ABSTRACTS OF JAPAN vol. 098, no. 001, 30 January 1998 & JP 09 235131 A (SHOWA ELECTRIC WIRE & CABLE CO LTD; NIPPON TELEGR & TELEPH CORP), 9 September 1997 see abstract ---	3,4
Y	PATENT ABSTRACTS OF JAPAN vol. 014, no. 227 (C-0718), 15 May 1990 & JP 02 055241 A (HITACHI CABLE LTD), 23 February 1990 see abstract ---	14,15
A	WO 97 21848 A (BAI WEI ;CHOY KWANG LEONG (GB); IMPERIAL COLLEGE (GB)) 19 June 1997 cited in the application ---	5
A	PATENT ABSTRACTS OF JAPAN vol. 013, no. 262 (C-608), 16 June 1989 & JP 01 065040 A (SUMITOMO ELECTRIC IND LTD), 10 March 1989 see abstract -----	13

# INTERNATIONAL SEARCH REPORT

Information on patent family members

International Application No

PCT/GB 98/03636

Patent document cited in search report	Publication date	Patent family member(s)	Publication date
WO 9721848 A	19-06-1997	GB 2308132 A	18-06-1997
		AU 1182997 A	03-07-1997
		CA 2240625 A	19-06-1997
		EP 0870075 A	14-10-1998
<hr/>			

## PATENT COOPERATION TREATY

## PCT

## INTERNATIONAL PRELIMINARY EXAMINATION REPORT



(PCT Article 36 and Rule 70)

Applicant's or agent's file reference W003952	<b>FOR FURTHER ACTION</b> See Notification of Transmittal of International Preliminary Examination Report (Form PCT/IPEA/416)	
International application No. PCT/GB98/03636	International filing date (day/month/year) 07/12/1998	Priority date (day/month/year) 05/12/1997
International Patent Classification (IPC) or national classification and IPC C23C16/44		
Applicant IMPERIAL COLLEGE OF SCIENCE, TECHNOLOGY ... et al.		

1. This international preliminary examination report has been prepared by this International Preliminary Examining Authority and is transmitted to the applicant according to Article 36.
2. This REPORT consists of a total of 6 sheets, including this cover sheet.
- ☒ This report is also accompanied by ANNEXES, i.e. sheets of the description, claims and/or drawings which have been amended and are the basis for this report and/or sheets containing rectifications made before this Authority (see Rule 70.16 and Section 607 of the Administrative Instructions under the PCT).
- These annexes consist of a total of 8 sheets.

3. This report contains indications relating to the following items:

- I ☒ Basis of the report
- II ☐ Priority
- III ☐ Non-establishment of opinion with regard to novelty, inventive step and industrial applicability
- IV ☐ Lack of unity of invention
- V ☒ Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement
- VI ☐ Certain documents cited
- VII ☐ Certain defects in the international application
- VIII ☒ Certain observations on the international application

Date of submission of the demand  23/06/1999	Date of completion of this report  23.03.00
Name and mailing address of the international preliminary examining authority:  European Patent Office D-80298 Munich Tel. +49 89 2399 - 0 Tx: 523656 epmu d Fax: +49 89 2399 - 4465	Authorized officer  Werner, H  Telephone No. +49 89 2399 8571 

**INTERNATIONAL PRELIMINARY  
EXAMINATION REPORT**

International application No. PCT/GB98/03636

**I. Basis of the report**

1. This report has been drawn on the basis of (*substitute sheets which have been furnished to the receiving Office in response to an invitation under Article 14 are referred to in this report as "originally filed" and are not annexed to the report since they do not contain amendments.*):

**Description, pages:**

1-5                      as received on                      14/02/2000    with letter of                      14/02/2000

**Claims, No.:**

1-28                      as received on                      14/02/2000    with letter of                      14/02/2000

**Drawings, sheets:**

1/1                      as originally filed

2. The amendments have resulted in the cancellation of:

- ☐ the description,              pages:  
☐ the claims,                  Nos.:  
☐ the drawings,                sheets:

3. ☒ This report has been established as if (some of) the amendments had not been made, since they have been considered to go beyond the disclosure as filed (Rule 70.2(c)):

~~see separate sheet~~

4. Additional observations, if necessary:

**INTERNATIONAL PRELIMINARY  
EXAMINATION REPORT**

International application No. PCT/GB98/03636

---

**V. Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement**

**1. Statement**

Novelty (N)	Yes:	Claims	4, 8, 10-12, 14, 19, 21-25
	No:	Claims	1, 5-6, 9, 13, 15-17, 26-28
Inventive step (IS)	Yes:	Claims	
	No:	Claims	1, 4-6, 8-17, 19, 21-28
Industrial applicability (IA)	Yes:	Claims	1, 4-6, 8-17, 19, 21-23
	No:	Claims	

**2. Citations and explanations**

**see separate sheet**

**VIII. Certain observations on the international application**

The following observations on the clarity of the claims, description, and drawings or on the question whether the claims are fully supported by the description, are made:

**see separate sheet**

1. SECTION I.3.

1.1. The amended claims 2, 3 (3 because it is dependent on claim 2), 7, 17, 18, and 20 as well as the description (page 2, line 5) filed with the letter dated 14.2.00, introduce subject-matter which extends beyond the content of the application as filed, contrary to Article 34(2)(b) PCT. The amendments concerned are the following:

- 1) "...an annular flow ... **about** the stream of droplets..." (in claims 2 and 7) and "...annular flow of..." (in claims 18 and 20). In the original application on page 2, lines 11-14 and Fig. 1 a **coaxial** nozzle assembly is disclosed. However, no *annular flow* has been disclosed in the original application.
- 2) "(the burner is) provided by the nozzle assembly" and "a fuel supply for supplying fuel to the nozzle assembly" (in claim 18). These features have not been disclosed in the original application.

Claims 2, 3, 7, 18, and 20 hence represent generalisations for which there is no basis in the original application. Therefore these claims are not considered regarding novelty, inventive step, and industrial applicability.

- 3) "a substrate holder for holding a substrate" (in claim 17 and page 2, line 5).

This feature has not been disclosed in the original application.

For the investigation regarding novelty, inventive step, and industrial applicability concerning claim 17 this feature was not considered.

2. SECTION V

2.1. Objections against Novelty (Art. 33(2) PCT):

2.1.1. Claims 1, 5, 6, 9, 13, 17, and 26 are not new according to Art. 33(2) PCT.

Document JP 56 005337 A discloses a method and an apparatus for depositing glass ( $\text{SiO}_2$ ) on a substrate comprising atomizing  $\text{Si}(\text{OC}_2\text{H}_5)_4$  to obtain a stream directed towards the substrate. An electrical field is applied between the nozzle and the target.  $\text{Si}(\text{OC}_2\text{H}_5)_4$  is mixed with hydrogen and the hydrogen is burned causing a flame directed towards the substrate and hence  $\text{Si}(\text{OC}_2\text{H}_5)_4$  is within the flame and thus being thermally decomposed before reaching the substrate. The  $\text{Si}(\text{OC}_2\text{H}_5)_4$  stream is divergent. A flow of an inert gas is delivered from the

nozzle towards the substrate. The substrate can be moved during the process of deposition.

- 2.1.2. Furthermore, also claims 15, 16, 27, and 28 are not new according to Art. 33(2) PCT since it is clear that the decomposition of  $\text{Si}(\text{OC}_2\text{H}_5)_4$  occurs in the flame, and hence away from or in the vicinity of the substrate.

2.2. Objections against inventive step (Art. 33(3) PCT):

- 2.2.1. Dependent claim 10, disclosing a multicomponent material, does not contain any features which, in combination with the features of any claim to which it refers, meet the requirements of the PCT in respect of inventive step (Art. 33(3) PCT), the reasons being as follows: Although document JP 56 005337 A does not disclose multicomponent materials, such materials are known to the person skilled in the art working in the area of semiconductors, and hence this feature is obvious.
- 2.2.2. Also the dependent claims 11 and 12 do not contain any features which, in combination with the features of any claim to which they refer, meet the requirements of the PCT in respect of inventive step, the reasons being as follows: Heating the substrate and using a sol precursor solution are two of several straightforward possibilities from which the skilled person would select, in accordance with circumstances, without the exercise of inventive skill, in order to solve the problem posed, and hence these features are obvious according to Art. 33(3) PCT.
- 2.2.3. Dependent claim 23 does not contain any features which, in combination with the features of any claim to which it refers, meet the requirements of the PCT in respect of inventive step (Art. 33(3) PCT), the reasons being as follows: Using a mesh does not seem to be of importance for solving the technical problem posed.
- 2.2.4. The features of dependent claims 24 and 25 have already been employed for the same purpose in a similar apparatus, see document JP 02 055241 A. It would therefore be obvious to the person skilled in the art to apply these features with corresponding effect to an apparatus as disclosed in JP 56 005337 A, thereby arriving at an apparatus according to claims 24 and 25.
- 2.2.5. Document JP 02 055241 A discloses an apparatus from which the subject-matter of claim 19 differs in that it discloses a nozzle assembly including an outlet for a



precursor **liquid** whereas in JP 02 055241 A the glass raw material is not precisely defined. However, the person skilled in the art knows that for such an apparatus the glass raw material must be a liquid or in the form of a fine powder. Furthermore, the person skilled in the art knows that in the present case the behaviour of a fine powder resembles a liquid. The glass raw material is reacted (hydrolysed) in the flame and the outlets are coaxial. Hence, the subject-matter of claim 19 does not involve an inventive step and does not satisfy the criterion set forth in Article 33(3) PCT.

- 2.2.6. The subject-matter of claims 21-22 does not involve an inventive step and does not satisfy the criterion set forth in Article 33(3) PCT because from document JP 02 055241 A taken in combination with document JP 56 005337 A the use of an extra outlet for cold gas is obvious. The difference to claims 21-22 is that in them the cold gas flow is between the precursor and the fuel whereas the combination of JP 56 005337 A and JP 02 055241 would result in a cold gas flow between the precursor-fuel and the oxygen. However, the technical effect would be the same, namely a possibility to control the combustion by the rate of cold gas.
- 2.2.7. For the same reasons as given in 2.2.5. the subject-matter of claim 4 does not involve an inventive step and does not satisfy the criterion set forth in Article 33(3) PCT.
- 2.2.8. For the same reasons as given in 2.2.6. the subject-matter of claim 8 does not involve an inventive step and does not satisfy the criterion set forth in Article 33(3) PCT.

### 3. SECTION VIII

#### 3.1. Clarity (Art. 6 PCT):

Claim 28 is written as a method claim. However, this seems to be erroneous because it refers back to apparatus claims. The investigation of claim 28 regarding novelty and inventive step has been performed assuming the claim is an apparatus claim.

09/555544

1 422 Rec'd PCT/PTO 31 JUN 2000

MATERIAL DEPOSITION

This invention relates to material deposition, for example, as a film or layer on a substrate or as a powder.

5 The application of materials such as ceramics as structural coatings and functional electronic films is expanding rapidly. Various desposition techniques such as chemical vapour deposition (CVD), physical vapour deposition (PVD), flame synthesis deposition (FSD), combustion chemical vapour deposition (CCVD) and sol-gel deposition have been developed or investigated.

10 Both CVD and PVD techniques involve the use of sophisticated and expensive deposition chambers and/or vacuum systems. The application of CVD and PVD techniques to deposit ceramic films is limited to coating processes in which the film thicknesses and coating areas are relatively small.

15 It is often difficult to control the stoichiometry of multicomponent oxide films deposited by CVD techniques, and problems can also arise due to differences in the vapour pressures of the CVD reagents and the low growth rate of CVD films.

PVD techniques such as radio frequency (RF) sputtering tend to give low deposition rates and poor yields, and reactive magnetic sputtering and ion beam sputtering need expensive equipment and skilled operators.

20 FSD techniques produce films with a morphology, microstructure and electrical properties which are dependent on the temperature of the substrate, the coating concentration, the carrier gas flow rate and so on. Control of all of these variables to achieve a desired coating is difficult.

25 Reference is also made to Hunt *et al*, Applied Physics 63 (1993), No 2, pages 266 to 268, WO-A-97/21848, GB-A-2192901, GB-A-2162861, EP-A-0103505, US-A-5652021, US-A-5534311 and SE-A-9504410.

This invention addresses these problems by providing a deposition technique which at least alleviates some of the disadvantages of the prior art.

30 This invention provides a method of depositing material on a substrate, comprising the steps of: delivering from an outlet a stream of droplets of a precursor liquid towards a substrate; applying an electric field between the outlet and the substrate; and generating a flame between the outlet and the substrate such that at least

a portion of the stream of droplets of the precursor liquid passes through the flame before reaching the substrate and the precursor liquid is chemically reacted and/or decomposed to provide the deposited material.

This invention also provides an apparatus for depositing material on a substrate, comprising: a substrate holder for holding a substrate; a nozzle assembly including an outlet from which a stream of droplets of a precursor liquid is in use delivered to a substrate; a precursor supply for supplying a precursor liquid to the nozzle assembly; an electrical supply for applying an electric field between the outlet and the substrate; and a burner for generating a flame between the outlet and the substrate and being configured such that in use at least a portion of the stream of droplets of the precursor liquid passes through the flame before reaching the substrate and the precursor liquid is chemically reacted and/or decomposed to provide the deposited material.

In a preferred embodiment the material is a ceramic material.

This invention provides a new technique which, in at least preferred embodiments, involves spraying atomised precursor droplets into a flame while providing an electric field between the precursor outlet and the substrate, so that the precursor forms a charged aerosol which undergoes combustion and/or chemical reaction in the vapour phase in the vicinity of the substrate and allows for the formation of a stable solid film with good adhesion to the substrate.

This invention will now be described, by way of example only, with reference to the accompanying drawings, throughout which like parts are described by like references, and in which Figure 1 is a schematic diagram of a deposition apparatus.

Figure 1 schematically illustrates a deposition apparatus comprising a coaxial nozzle assembly 10 having a liquid precursor delivery capillary 20, a first coaxial passage 30 for cold air, nitrogen or other gases, and a second coaxial passage 40 for liquid or gaseous fuel.

The precursor can be, for example, one of the precursors listed in WO-A-97/21848, with or without the mentioned catalyst. Many other precursors can be used as appropriate for the desired deposition, such as precursors known from FSD techniques as disclosed by Choy in "Flame Assisted Vapour Deposition of Ceramic

Films and Coatings", British Ceramic Proceedings, The Institute of Materials (1995), pages 65 to 74.

The fuel may be a mixture of oxygen and acetylene, or another appropriate fuel, such as fuels known from FSD techniques.

5 A high voltage source 45 maintains an electric field between the nozzle assembly 10 and a substrate 50. The potential difference may be, for example, within the approximate ranges described in WO-A-97/21848, typically in the approximate range of from 5 to 30 kV.

10 The precursor liquid is sprayed towards a region 55 of the substrate 50 from an outlet 60 of the capillary 20. The fuel is ignited so that an annular combustion region 70 is generated. The extent of this combustion region can be controlled by controlling the fuel flow rate, the distance between the nozzle assembly 10 and the substrate 50, the amount and flow rate of cold gases in the passage 30, and the applied electric field.

15 Decomposition and/or chemical reaction of the precursor, for example, a sol-gel transition, occurs in a higher temperature overlap zone between the spray of precursor from the outlet 60 of the capillary 20 and the combustion region 70. Deposition occurs in or beneath this overlap zone. So, by controlling the extent of the combustion region as described above, the deposition on the substrate 50 can be controlled, and premature reaction or decomposition, which is a problem in many prior art FSD techniques leading to non-uniform deposition, can be avoided.

20 Either polarity of electric field can be used, ~~or a periodically or occasionally~~ alternating field can be employed. A thermocouple can be used to monitor the temperature of the substrate 50.

25 The apparatus preferably includes a mesh 90 which assists in removing soot from the flame and so provide a high temperature (blue) flame.

The apparatus preferably includes a further annular electrode 100 connected to the high voltage supply 45 at an intermediate potential between that of the nozzle assembly 10 and the substrate 50 to steer the material being deposited onto a required area of the substrate 50.

30 The technique is also applicable to premixed fuel and precursor systems. However, non-premixed systems are preferred as these systems give greater control of the deposition temperature and assist in avoiding premature decomposition.

The technique can be used to manufacture metal oxide and non-oxide materials; to manufacture pure, doped, multicomponent or multiphase materials; to manufacture materials with dense, porous, or a combination of dense and porous structures; to manufacture composite, multilayer and compositionally-graded structures; to produce thin or thick films; for rapid prototyping of net shape and near net shape components; or to coat planar or tubular substrates or other complex shaped components.

The technique can be scaled up for large area or mass production by using multiple flame/electrostatic units. For accurate deposition and process control, the process can be automated.

The substrate 50 can be conductive or non-conductive. For non-conductive substrates, the conductivity can be improved by utilizing a conductive backing holder.

The technique can be performed in an open atmosphere or in an inert/controlled atmosphere. For example, oxide-based structures can be deposited in an open atmosphere, and non-oxide structures, such as sulphides, carbides, etc, can be deposited in a controlled atmosphere. Deposition can take place at atmospheric or a different pressure.

The chemistry of the precursors can be adjusted so that once the chemical reaction starts to take place, a self-assisted reaction occurs. This can reduce the requirements for fuel, while still achieving the required deposition temperature for a particular material.

The electric field reduces the loss of precursor to the surroundings by directing the precursor to the deposition surface. This is a clear advantage over conventional flame-based techniques.

The deposition can be controlled by one or more of the following: the flow rate of the cold gas; the electric field strength; the fuel and its flow rate; the separation of the nozzle assembly from the substrate; the chemistry, concentration and flow rate of the precursor; and the deposition pressure.

Embodiments of the invention allow the use of simple, flexible and/or mobile equipment. The technique can be made relatively safe by the use of sol precursors and/or water based precursors. The process can give rise to an advantageously low flame/deposition temperature for crystalline materials, for example from 550 to 800 °C

for  $Y_2O_3-ZrO_2$ . Dense films tend to require a sol precursor, whereas porous films may be based on sol or water based precursors. The consumption of precursor can be relatively low, for example, 1 ml of 0.05 M solution to generate a 1  $\mu m$  film measuring 1 cm x 1 cm. Furthermore, the deposition can be performed in a single step without  
5 the need for a subsequent heat treatment.

Powders can be formed by providing for the chemical reaction of the precursor to the solid phase to take place above the substrate 50. With this configuration, the substrate 50 is deposited with discrete powder particles which can be later collected. Powder generation can be improved by employing gas condensation techniques and a  
10 cooled collecting substrate.

In a further embodiment, the substrate 50 can be mounted on a movable table or XY positioner under the control of, for example, a computer aided design (CAD) system to allow three-dimensional objects to be constructed layer by layer. This can be used in, for example, rapid prototyping systems.

CLAIMS

1. A method of depositing material on a substrate, comprising the steps of:  
delivering from an outlet a stream of droplets of a precursor liquid towards a  
substrate;  
5 applying an electric field between the outlet and the substrate; and  
generating a flame between the outlet and the substrate such that at least a  
portion of the stream of droplets of the precursor liquid passes through the  
flame before reaching the substrate and the precursor liquid is chemically  
reacted and/or decomposed to provide the deposited material.  
10
2. The method according to claim 1, wherein the flame generation step comprises  
the step of delivering from a second outlet an annular flow of fuel about the  
stream of droplets such as to provide an annular flame combustion region  
through which at least the portion of the stream of droplets passes before  
15 reaching the substrate.
3. The method according to claim 2, wherein the annular flow of fuel is a  
diverging flow.
- 20 4. The method according to claim 2 or 3, wherein the first and second outlets are  
coaxial.
5. The method according to any of claims 1 to 4, wherein the stream of droplets is  
provided as a diverging spray.  
25
6. The method according to any of claims 1 to 5, further comprising the step of  
delivering a flow of cold gas in the direction from the first outlet towards the  
substrate.

7. The method according to claim 6 when appendant upon claim 2, wherein the flow of cold gas is delivered from a third outlet as an annular flow about the stream of droplets and within the annular flow of fuel.
- 5 8. The method according to claim 7, wherein the first and third outlets are coaxial.
9. The method according to any of claims 1 to 8, wherein the material is a ceramic material.
- 10 10. The method according to any of claims 1 to 9, wherein the material is a multicomponent oxide material.
11. The method according to any of claims 1 to 10, further comprising the step of heating the substrate.
- 15 12. The method according to any of claims 1 to 11, wherein the precursor liquid is a sol precursor solution.
13. The method according to any of claims 1 to 12, further comprising the step of moving one or both of the substrate and the first outlet during deposition so as to deposit a three-dimensional structure as a series of overlapping layers.
- 20 14. The method according to any of claims 1 to 13, further comprising the step of controlling the region of deposition by varying one or more of the rate of flow of the fuel, the separation between the first outlet and the substrate and the electric field between the first outlet and the substrate.
- 25 15. The method according to any of claims 1 to 14, wherein the material is deposited as a powder and the chemical reaction and/or decomposition occurs away from the substrate.
- 30



16. The method according to any of claims 1 to 14, wherein the material is deposited as a solid film and the chemical reaction and/or decomposition occurs in the vicinity of the substrate.
- 5 17. An apparatus for depositing material on a substrate, comprising:  
a substrate holder for holding a substrate;  
a nozzle assembly including an outlet from which a stream of droplets of a precursor liquid is in use delivered to a substrate;  
a precursor supply for supplying a precursor liquid to the nozzle assembly;  
10 an electrical supply for applying an electric field between the outlet and the substrate; and  
a burner for generating a flame between the outlet and the substrate and being configured such that in use at least a portion of the stream of droplets of the precursor liquid passes through the flame before reaching the substrate and the  
15 precursor liquid is chemically reacted and/or decomposed to provide the deposited material.
18. The apparatus according to claim 17, wherein the burner is provided by the  
20 nozzle assembly and the nozzle assembly includes a second outlet from which an annular flow of fuel is in use delivered ~~such as to provide an annular flame~~ combustion region through which at least the portion of the stream of droplets passes before reaching the substrate, and further comprising a fuel supply for supplying fuel to the nozzle assembly.
- 25 19. The apparatus according to claim 18, wherein the first and second outlets are coaxial.
20. The apparatus according to claim 18 or 19, wherein the nozzle assembly further  
30 comprises a third outlet disposed between the first and second outlets from which an annular flow of cold gas is in use delivered.

21. The apparatus according to claim 20, wherein the first and third outlets are coaxial.
22. The apparatus according to any of claims 18 to 21, wherein the first outlet is a central outlet.
23. The apparatus according to any of claims 17 to 22, further comprising a mesh disposed between the first outlet and the substrate.
24. The apparatus according to any of claims 17 to 23, further comprising an electrode at an electric potential between the potential of the first outlet and the substrate and disposed between the first outlet and the substrate.
25. The apparatus according to claim 24, wherein the electrode is an annular electrode.
26. The apparatus according to any of claims 17 to 25, further comprising a positioner for altering the relative position of the first outlet and the substrate.
27. The apparatus according to any of claims 17 to 26, where configured such that the chemical reaction and/or decomposition occurs away from the substrate so as to provide the material as a powder.
28. The method according to any of claims 17 to 26, where configured such that the chemical reaction and/or decomposition occurs in the vicinity of the substrate so as to provide the material as a solid film.

PCT

NOTICE INFORMING THE APPLICANT OF THE  
COMMUNICATION OF THE INTERNATIONAL  
APPLICATION TO THE DESIGNATED OFFICES

(PCT Rule 47.1(c), first sentence)

From the INTERNATIONAL BUREAU

To:

TURNER, James, Arthur  
D. Young & Co.  
21 New Fetter Lane  
London EC4A 1DA  
ROYAUME-UNICOMPUTER  
NOTED

JATS

Date of mailing (day/month/year)

17 June 1999 (17.06.99)

Applicant's or agent's file reference

P/3952.WO

## IMPORTANT NOTICE

International application No.

PCT/GB98/03636

International filing date (day/month/year)

07 December 1998 (07.12.98)

Priority date (day/month/year)

05 December 1997 (05.12.97)

Applicant

IMPERIAL COLLEGE OF SCIENCE, TECHNOLOGY AND MEDICINE et al

1. Notice is hereby given that the International Bureau has communicated, as provided in Article 20, the international application to the following designated Offices on the date indicated above as the date of mailing of this Notice:

AU,CN,EP,IL,JP,KP,KR,US

In accordance with Rule 47.1(c), third sentence, those Offices will accept the present Notice as conclusive evidence that the communication of the international application has duly taken place on the date of mailing indicated above and no copy of the international application is required to be furnished by the applicant to the designated Office(s).

2. The following designated Offices have waived the requirement for such a communication at this time:

AL,AM,AP,AT,AZ,BA,BB,BG,BR,BY,CA,CH,CU,CZ,DE,DK,EA,EE,ES,FI,GB,GD,GE,GH,GM,HR,HU,  
ID,IN,IS,KE,KG,KZ,LC,LK,LR,LS,LT,LU,LV,MD,MG,MK,MN,MW,MX,NO,NZ,OA,PL,PT,RO,RU,SD,  
SE,SG,SI,SK,SL,TJ,TM,TR,TT,UA,UG,UZ,VN,YU,ZW

The communication will be made to those Offices only upon their request. Furthermore, those Offices do not require the applicant to furnish a copy of the international application (Rule 49.1(a-bis)).

3. Enclosed with this Notice is a copy of the international application as published by the International Bureau on

17 June 1999 (17.06.99), under No. P/3952.WO

## REMINDER REGARDING CHAPTER II (Article 31(2)(a) and Rule 54.2)

If the applicant wishes to postpone entry into the national phase until 30 months (or later in some Offices) from the priority date, a **demand for international preliminary examination** must be filed with the competent International Preliminary Examining Authority before the expiration of 19 months from the priority date.

It is the applicant's sole responsibility to monitor the 19-month time limit.

Note that only an applicant who is a national or resident of a PCT Contracting State which is bound by Chapter II has the right to file a demand for international preliminary examination.

## REMINDER REGARDING ENTRY INTO THE NATIONAL PHASE (Article 22 or 39(1))

If the applicant wishes to proceed with the international application in the **national phase**, he must, within 20 months or 30 months, or later in some Offices, perform the acts referred to therein before each designated or elected Office.

For further important information on the time limits and acts to be performed for entering the national phase, see the Annex to Form PCT/IB/301 (Notification of Receipt of Record Copy) and Volume II of the PCT Applicant's Guide.

The International Bureau of WIPO  
34, chemin des Colombettes  
1211 Geneva 20, Switzerland

Facsimile No. (41-22) 740.14.35

Authorized officer

J. Zahra

Telephone No. (41-22) 338.83.38

**NOTICE INFORMING THE APPLICANT OF THE COMMUNICATION OF  
THE INTERNATIONAL APPLICATION TO THE DESIGNATED OFFICES**

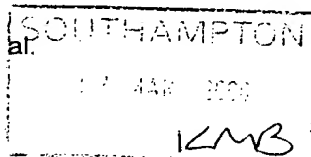
<b>Date of mailing (day/month/year)</b> 17 June 1999 (17.06.99)	<b>IMPORTANT NOTICE</b>
<b>Applicant's or agent's file reference</b> P/3952.WO	<b>International application No.</b> PCT/GB98/03636
<p>The applicant is hereby notified that, at the time of establishment of this Notice, the time limit under Rule 46.1 for making amendments under Article 19 has not yet expired and the International Bureau had received neither such amendments nor a declaration that the applicant does not wish to make amendments.</p>	

# PATENT COOPERATION TREATY

From the  
INTERNATIONAL PRELIMINARY EXAMINING AUTHORITY

To:

TURNER, James Arthur et al.  
D YOUNG & CO  
21 New Fetter Lane  
London EC4A 1DA  
GRANDE BRETAGNE



## PCT

### NOTIFICATION OF TRANSMITTAL OF THE INTERNATIONAL PRELIMINARY EXAMINATION REPORT

(PCT Rule 71.1)

Date of mailing  
(day/month/year)

2 3. 03. 00

Applicant's or agent's file reference  
W003952

#### IMPORTANT NOTIFICATION

International application No.  
PCT/GB98/03636

International filing date (day/month/year)  
07/12/1998

Priority date (day/month/year)  
05/12/1997

Applicant  
IMPERIAL COLLEGE OF SCIENCE, TECHNOLOGY ... et al.

1. The applicant is hereby notified that this International Preliminary Examining Authority transmits herewith the international preliminary examination report and its annexes, if any, established on the international application.
2. A copy of the report and its annexes, if any, is being transmitted to the International Bureau for communication to all the elected Offices.
3. Where required by any of the elected Offices, the International Bureau will prepare an English translation of the report (but not of any annexes) and will transmit such translation to those Offices.

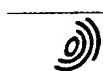
#### 4. REMINDER

The applicant must enter the national phase before each elected Office by performing certain acts (filing translations and paying national fees) within 30 months from the priority date (or later in some Offices) (Article 39(1)) (see also the reminder sent by the International Bureau with Form PCT/IB/301).

Where a translation of the international application must be furnished to an elected Office, that translation must contain a translation of any annexes to the international preliminary examination report. It is the applicant's responsibility to prepare and furnish such translation directly to each elected Office concerned.

For further details on the applicable time limits and requirements of the elected Offices, see Volume II of the PCT Applicant's Guide.

Name and mailing address of the IPEA/



European Patent Office  
D-80298 Munich  
Tel. +49 89 2399 - 0 Tx: 523656 epmu d  
Fax: +49 89 2399 - 4465

Authorized officer

Borinski, W

Tel. +49 89 2399-8237





## REQUEST

The undersigned requests that the present international application be processed according to the Patent Cooperation Treaty.

For receiving office use only

International Application No.

International Filing Date

Name of receiving Office and "PCT International Application"

Applicant's or agent's file reference  
(if desired) (12 characters maximum)

P/3952.WO

<b>Box No. I</b>	<b>TITLE OF INVENTION</b>	<b>MATERIAL DEPOSITION</b>
<b>Box No. II APPLICANT</b>		
Name and address: (Family name followed by given name; for a legal entity, full official designation. The address must include postal code and name of country. The country of the address indicated in this Box is the applicant's State (i.e. country) of residence if no State of residence is indicated below.)		<input type="checkbox"/> This person is also inventor.
IMPERIAL COLLEGE OF SCIENCE, TECHNOLOGY AND MEDICINE Sherfield Building Exhibition Road London SW7 2AZ United Kingdom		Telephone No.
		Facsimile No.
		Teleprinter No.
State (i.e. country) of nationality: United Kingdom		State (i.e. country) of residence: United Kingdom
This person is applicant for the purposes of: <input type="checkbox"/> all designated States <input checked="" type="checkbox"/> all designated States except the United States of America <input type="checkbox"/> the United States of America only <input type="checkbox"/> the States indicated in the Supplemental Box		
<b>Box No. III FURTHER APPLICANT(S) AND/OR (FURTHER) INVENTOR(S)</b>		
Name and address: (Family name followed by given name; for a legal entity, full official designation. The address must include postal code and name of country. The country of the address indicated in this Box is the applicant's State (i.e. country) of residence if no State of residence is indicated below.)		This person is:
CHOY, Kwang-Leong 35 Blondvil Street Cheylesmeore Coventry CB3 5EQ United Kingdom		<input type="checkbox"/> applicant only
		<input checked="" type="checkbox"/> applicant and inventor
		<input type="checkbox"/> inventor only (if this check-box is marked, do not fill in below)
State (i.e. country) of nationality: <del>United Kingdom</del>		State (i.e. country) of residence: <del>United Kingdom</del>
This person is applicant for the purposes of: <input type="checkbox"/> all designated States <input type="checkbox"/> all designated States except the United States of America <input checked="" type="checkbox"/> the United States of America only <input type="checkbox"/> the States indicated in the Supplemental Box		
<input checked="" type="checkbox"/> Further applicant and/or (further) inventors are indicated on a continuation sheet		
<b>Box No. IV AGENT OR COMMON REPRESENTATIVE; OR ADDRESS FOR CORRESPONDENCE</b>		
The person identified below is hereby/has been appointed to act on behalf of the applicant(s) before the competent International Authorities as: <input checked="" type="checkbox"/> agent <input type="checkbox"/> common representative		
Name and address: (Family name followed by given name; for a legal entity, full official designation. The address must include postal code and name of country.)		Telephone No.
TURNER, James Arthur et al D Young & Co 21 New Fetter Lane London EC4A 1DA		+44 1703 634816
		Facsimile No.
		+44 1703 224262
		Teleprinter No.
		477667 YOUNGS G
Mark this check-box where no agent or common representative is/has been appointed and the space above is used instead to indicate a special address to which correspondence should be sent.		

Continuation of Box No. III		FURTHER APPLICANTS AND/OR (FURTHER) INVENTORS	
<i>If none of the following sub-boxes is used, this sheet is not to be included in the request.</i>			
Name and address: (Family name followed by given name; for a legal entity, full official designation. The address must include postal code and name of country. The country of the address indicated in this Box is the applicant's State (i.e. country) of residence if no State of residence is indicated below.)  CHANG, Isaac Tsz Hong c/o School of Metallurgy and Materials University of Birmingham Edgbaston Birmingham B15 2TT United Kingdom		This person is:  <input type="checkbox"/> applicant only  <input checked="" type="checkbox"/> applicant and inventor  <input type="checkbox"/> inventor only (if this check-box is marked, do not fill in below)	
State (i.e. country) of nationality: United Kingdom		State (i.e. country) of residence: United Kingdom	
This person is applicant for the purposes of: <input type="checkbox"/> all designated States <input type="checkbox"/> all designated States except the United States of America <input checked="" type="checkbox"/> the United States of America only <input type="checkbox"/> the States indicated in the Supplemental Box			
Name and address: (Family name followed by given name; for a legal entity, full official designation. The address must include postal code and name of country. The country of the address indicated in this Box is the applicant's State (i.e. country) of residence if no State of residence is indicated below.)		This person is:  <input type="checkbox"/> applicant only  <input type="checkbox"/> applicant and inventor  <input type="checkbox"/> inventor only (if this check-box is marked, do not fill in below)	
State (i.e. country) of nationality:		State (i.e. country) of residence:	
This person is applicant for the purposes of: <input type="checkbox"/> all designated States <input type="checkbox"/> all designated States except the United States of America <input type="checkbox"/> the United States of America only <input type="checkbox"/> the States indicated in the Supplemental Box			
Name and address: (Family name followed by given name; for a legal entity, full official designation. The address must include postal code and name of country. The country of the address indicated in this Box is the applicant's State (i.e. country) of residence if no State of residence is indicated below.)		This person is:  <input type="checkbox"/> applicant only  <input type="checkbox"/> applicant and inventor  <input type="checkbox"/> inventor only (if this check-box is marked, do not fill in below)	
State (i.e. country) of nationality:		State (i.e. country) of residence:	
This person is applicant for the purposes of: <input type="checkbox"/> all designated States <input type="checkbox"/> all designated States except the United States of America <input type="checkbox"/> the United States of America only <input type="checkbox"/> the States indicated in the Supplemental Box			
Name and address: (Family name followed by given name; for a legal entity, full official designation. The address must include postal code and name of country. The country of the address indicated in this Box is the applicant's State (i.e. country) of residence if no State of residence is indicated below.)		This person is:  <input type="checkbox"/> applicant only  <input type="checkbox"/> applicant and inventor  <input type="checkbox"/> inventor only (if this check-box is marked, do not fill in below)	
State (i.e. country) of nationality:		State (i.e. country) of residence:	
This person is applicant for the purposes of: <input type="checkbox"/> all designated States <input type="checkbox"/> all designated States except the United States of America <input type="checkbox"/> the United States of America only <input type="checkbox"/> the States indicated in the Supplemental Box			
Name and address: (Family name followed by given name; for a legal entity, full official designation. The address must include postal code and name of country. The country of the address indicated in this Box is the applicant's State (i.e. country) of residence if no State of residence is indicated below.)			
State (i.e. country) of nationality:		State (i.e. country) of residence:	
This person is applicant for the purposes of: <input type="checkbox"/> all designated States <input type="checkbox"/> all designated States except the United States of America <input type="checkbox"/> the United States of America only <input type="checkbox"/> the States indicated in the Supplemental Box			
<input type="checkbox"/> Further applicants and/or (further) inventors are indicated on a continuation sheet			

**Box No. V DESIGNATION OF STATES**

The following designations are hereby made under Rule 4.9(a) (mark the applicable check-boxes; at least one must be marked):

**Regional Patent**

- ☒ **AP** **ARIPO Patent:** GH Ghana, GM Gambia, KE Kenya, LS Lesotho, MW Malawi, SD Sudan, SZ Swaziland, UG Uganda, ZW Zimbabwe, and any other State which is a Contracting State of the Harare Protocol and of the PCT
- ☒ **EA** **Eurasian Patent:** AM Armenia, AZ Azerbaijan, BY Belarus, KG Kyrgyzstan, KZ Kazakstan, MD Republic of Moldova, RU Russian Federation, TJ Tajikistan, TM Turkmenistan, and any other State which is a Contracting State of the Eurasian Patent Convention and of the PCT
- ☒ **EP** **European Patent:** AT Austria, BE Belgium, CH and LI Switzerland and Liechtenstein, CY Cyprus, DE Germany, DK Denmark, ES Spain, FI Finland, FR France, GB United Kingdom, GR Greece, IE Ireland, IT Italy, LU Luxembourg, MC Monaco, NL Netherlands, PT Portugal, SE Sweden, and any other State which is a Contracting State of the European Patent Convention and of the PCT
- ☒ **OA** **OAPI Patent:** BF Burkina Faso, BJ Benin, CF Central African Republic, CG Congo, CI Côte d'Ivoire, CM Cameroon, GA Gabon, GN Guinea, ML Mali, MR Mauritania, NE Niger, SN Senegal, TD Chad, TG Togo, and any other State which is a member State of OAPI and a Contracting State of the PCT (if any other kind of protection or treatment desired, please specify on dotted line) .....

**National Patent (if other kind of protection or treatment desired, specify on dotted line):**

- |   |  |
|---|--|
| <input checked="" type="checkbox"/> <b>AL</b> Albania                               | <input checked="" type="checkbox"/> <b>LT</b> Lithuania  |
| <input checked="" type="checkbox"/> <b>AM</b> Armenia                               | <input checked="" type="checkbox"/> <b>LU</b> Luxembourg   |
| <input checked="" type="checkbox"/> <b>AT</b> Austria                               | <input checked="" type="checkbox"/> <b>LV</b> Latvia   |
| <input checked="" type="checkbox"/> <b>AU</b> Australia                             | <input checked="" type="checkbox"/> <b>MD</b> Republic of Moldova  |
| <input checked="" type="checkbox"/> <b>AZ</b> Azerbaijan                            | <input checked="" type="checkbox"/> <b>MG</b> Madagascar   |
| <input checked="" type="checkbox"/> <b>BA</b> Bosnia and Herzegovina                | <input checked="" type="checkbox"/> <b>MK</b> The former Yugoslav Republic of Macedonia  |
| <input checked="" type="checkbox"/> <b>BB</b> Barbados                              | <input checked="" type="checkbox"/> <b>MN</b> Mongolia   |
| <input checked="" type="checkbox"/> <b>BG</b> Bulgaria                              | <input checked="" type="checkbox"/> <b>MW</b> Malawi   |
| <input checked="" type="checkbox"/> <b>BR</b> Brazil                                | <input checked="" type="checkbox"/> <b>MX</b> Mexico   |
| <input checked="" type="checkbox"/> <b>BY</b> Belarus                               | <input checked="" type="checkbox"/> <b>NO</b> Norway   |
| <input checked="" type="checkbox"/> <b>CA</b> Canada                                | <input checked="" type="checkbox"/> <b>NZ</b> New Zealand  |
| <input checked="" type="checkbox"/> <b>CH AND LI</b> Switzerland and Liechtenstein  | <input checked="" type="checkbox"/> <b>PL</b> Poland   |
| <input checked="" type="checkbox"/> <b>CN</b> China                                 | <input checked="" type="checkbox"/> <b>PT</b> Portugal   |
| <input checked="" type="checkbox"/> <b>CU</b> Cuba                                  | <input checked="" type="checkbox"/> <b>RO</b> Romania  |
| <input checked="" type="checkbox"/> <b>CZ</b> Czech Republic                        | <input checked="" type="checkbox"/> <b>RU</b> Russian Federation   |
| <input checked="" type="checkbox"/> <b>DE</b> Germany                               | <input checked="" type="checkbox"/> <b>SD</b> Sudan  |
| <input checked="" type="checkbox"/> <b>DK</b> Denmark                               | <input checked="" type="checkbox"/> <b>SE</b> Sweden   |
| <input checked="" type="checkbox"/> <b>EE</b> Estonia                               | <input checked="" type="checkbox"/> <b>SG</b> Singapore  |
| <input checked="" type="checkbox"/> <b>ES</b> Spain                                 | <input checked="" type="checkbox"/> <b>SI</b> Slovenia   |
| <input checked="" type="checkbox"/> <b>FI</b> Finland                               | <input checked="" type="checkbox"/> <b>SK</b> Slovakia   |
| <input checked="" type="checkbox"/> <b>GB</b> United Kingdom                        | <input checked="" type="checkbox"/> <b>SL</b> Sierra Leone   |
| <input checked="" type="checkbox"/> <b>GE</b> Georgia                               | <input checked="" type="checkbox"/> <b>TJ</b> Tajikistan   |
| <input checked="" type="checkbox"/> <b>GH</b> Ghana                                 | <input checked="" type="checkbox"/> <b>TM</b> Turkmenistan   |
| <input checked="" type="checkbox"/> <b>GM</b> Gambia                                | <input checked="" type="checkbox"/> <b>TR</b> Turkey   |
| <input checked="" type="checkbox"/> <b>GW</b> Guinea-Bissau                         | <input checked="" type="checkbox"/> <b>TT</b> Trinidad and Tobago  |
| <input checked="" type="checkbox"/> <b>HU</b> Hungary                               | <input checked="" type="checkbox"/> <b>UA</b> Ukraine  |
| <input checked="" type="checkbox"/> <b>ID</b> Indonesia                             | <input checked="" type="checkbox"/> <b>UG</b> Uganda   |
| <input checked="" type="checkbox"/> <b>IL</b> Israel                                | <input checked="" type="checkbox"/> <b>US</b> United States of America   |
| <input checked="" type="checkbox"/> <b>IS</b> Iceland                               |  |
| <input checked="" type="checkbox"/> <b>JP</b> Japan                                 |  |
| <input checked="" type="checkbox"/> <b>KE</b> Kenya                                 | <input checked="" type="checkbox"/> <b>UZ</b> Uzbekistan   |
| <input checked="" type="checkbox"/> <b>KG</b> Kyrgyzstan                            | <input checked="" type="checkbox"/> <b>VN</b> Viet Nam   |
| <input checked="" type="checkbox"/> <b>KP</b> Democratic People's Republic of Korea | <input checked="" type="checkbox"/> <b>YU</b> Yugoslavia   |
|   | <input checked="" type="checkbox"/> <b>ZW</b> Zimbabwe   |
| <input checked="" type="checkbox"/> <b>KR</b> Republic of Korea                     | Check-boxes reserved for designating States (for the purposes of a national patent) which have become party to the PCT after the issuance of this sheet: |
| <input checked="" type="checkbox"/> <b>KZ</b> Kazakstan                             | <input checked="" type="checkbox"/> <b>HR</b> Croatia  |
| <input checked="" type="checkbox"/> <b>LC</b> Saint Lucia                           | <input checked="" type="checkbox"/> <b>GD</b> Grenada  |
| <input checked="" type="checkbox"/> <b>LK</b> Sri Lanka                             | <input checked="" type="checkbox"/> <b>IN</b> India  |
| <input checked="" type="checkbox"/> <b>LR</b> Liberia                               |  |
| <input checked="" type="checkbox"/> <b>LS</b> Lesotho                               |  |

In addition to the designations made above, the applicant also makes under Rule 4.9(b) all designations which would be permitted under the PCT except the designation(s) of

The applicant declares that those additional designations are subject to confirmation and that any designation which is not confirmed before the expiration of 15 months from the priority date is to be regarded as withdrawn by the applicant at the expiration of that time limit. (Confirmation of a designation consists of the filing of a notice specifying that designation and the payment of the designation and confirmation fees. Confirmation must reach the receiving Office within the 15-month time limit.)



<b>Box No. VI PRIORITY CLAIM</b>		Further priority claims are indicated in the Supplemental Box <input type="checkbox"/>	
The priority of the following earlier application(s) is hereby claimed:			
Country (in which, or for which, the application was filed)	Filing Date (day/month/year)	Application No.	Office of filing (only for regional or international application)
item (1) United Kingdom	5 Dec 1997	9725878.4	
item (2)			
item (3)			
Mark the following check-box if the certified copy of the earlier application is to be issued by the Office which for the purposes of the present international application is the receiving Office (a fee may be required):			
<input checked="" type="checkbox"/> The receiving Office is hereby requested to prepare and transmit to the International Bureau a certified copy of the earlier application(s) identified above as item(s) : (1)			
<b>Box No. VII INTERNATIONAL SEARCHING AUTHORITY</b>			
Choice of International Searching Authority (ISA) (If two or more International Searching Authorities are competent to carry out the international search, indicate the Authority chosen; the two-letter code may be used): ISA /			
Earlier Search Fill in where a search (international, international-type or other) by the International Search Authority has already been carried out or requested and the Authority is now requested to base the international search, to the extent possible, on the results of that earlier search. Identify such search or request either by reference to the relevant application (or the translation thereof) or by reference to the search request:			
Country (or regional Office):	Date (day/month/year)	Number:	
<b>Box No. VII CHECK LIST</b>			
This international application contains the following number of sheets:		This international application is accompanied by the item(s) marked below:	
1. request : 4 sheets		1. <input type="checkbox"/> separate signed power of attorney	5. <input checked="" type="checkbox"/> fee calculation sheet
2. description : 5 sheets		2. <input checked="" type="checkbox"/> copy of general power of attorney	6. <input type="checkbox"/> separate indications concerning deposited microorganisms
3. claims : 3 sheets		3. <input type="checkbox"/> statement explaining lack of signature	7. <input type="checkbox"/> nucleotide and/or amino acid sequence listing (diskette)
4. abstract : 1 sheets		4. <input type="checkbox"/> priority documents(s) identified in Box No. VI as item(s):	8. <input type="checkbox"/> other (specify):
5. drawings : 1 sheets			
Total : 14 sheets			
Figure No. 1 of the drawings (if any) should accompany the abstract when it is published			
<b>Box No. IX SIGNATURE OF APPLICANT OR AGENT</b>			
Next to each signature, indicate the name of the person signing and the capacity in which the person signs (if such capacity is not obvious from reading the request)			

For receiving Office use only	
1. Date of actual receipt of the purported international application:	2. Drawings:
3. Corrected date of actual receipt due to later but timely received papers or drawings completing the purported international application:	<input type="checkbox"/> received:
4. Date of timely receipt of the required corrections under PCT Article 11(2):	<input type="checkbox"/> not received:
5. International Searching Authority specified by the applicant: ISA /	6. <input type="checkbox"/> Transmittal of search copy delayed until search fee paid

For International Bureau use only	
Date of receipt of the record copy by the International Bureau:	

From the RECEIVING OFFICE

PCT

NOTIFICATION OF THE INTERNATIONAL  
APPLICATION NUMBER AND OF THE  
INTERNATIONAL FILING DATE

(PCT Rule 20.5(c))

To: D Young & Co 21 New Fetter Lane London  EC4A 1DA
---

Date of mailing  
(day/month/year)

14 DECEMBER 1998

Applicant's or agents's file reference  
P/3952.WO

## IMPORTANT NOTIFICATION

International application No.  
PCT/GB98/03636International filing date (day/month/year)  
6/7/12/1998Priority date (day/month/year)  
05/12/1997Applicant  
Imperial College of Science, Technology and Medicine et alTitle of the invention  
Material deposition

1. The applicant is hereby notified that the international application has been accorded the international application number and the international filing date indicated above.

2. The applicant is further notified that the record copy of the international application:



was transmitted to the International Bureau on

14 DECEMBER 1998



has not yet been transmitted to the International Bureau for the reason indicated below and a copy of this notification has been sent to the International Bureau\*:



because the necessary national security clearance has not yet been obtained.



because (reason to be specified):

\* The International Bureau monitors the transmittal of the record copy by the receiving Office and will notify the applicant (with Form PCT/IB/301) of its receipt. Should the record copy not have been received by the expiration of 14 months from the priority date, the International Bureau will notify the applicant (Rule 22.1(c)).

Name and mailing address of the receiving Office The Patent Office Cardiff Road, Newport South Wales NP9 1RH  Facsimile No.	Authorized officer  Steve Bevan  Telephone No. 01633 814588
--	---

## PATENT COOPERATION TREATY

PCT

From the INTERNATIONAL BUREAU

NOTIFICATION OF RECEIPT OF  
RECORD COPY

(PCT Rule 24.2(a))

To:

TURNER, James, Arthur  
D Young & Co  
21 New Fetter Lane  
London EC4A 1DA  
ROYAUME-UNI

Date of mailing (day/month/year) 04 January 1999 (04.01.99)	<b>IMPORTANT NOTIFICATION</b>
Applicant's or agent's file reference P/3952.WO	International application No. PCT/GB98/03636

The applicant is hereby notified that the International Bureau has received the record copy of the international application as detailed below.

Name(s) of the applicant(s) and State(s) for which they are applicants:

IMPERIAL COLLEGE OF SCIENCE, TECHNOLOGY AND MEDICINE (for all designated States except US)

CHOY, Kwang-Leong et al (for US)

International filing date : 07 December 1998 (07.12.98)

Priority date(s) claimed : 05 December 1997 (05.12.97)

Date of receipt of the record copy by the International Bureau : 22 December 1998 (22.12.98)

List of designated Offices :

AP : GH, GM, KE, LS, MW, SD, SZ, UG, ZW

EA : AM, AZ, BY, KG, KZ, MD, RU, TJ, TM

EP : AT, BE, CH, CY, DE, DK, ES, FI, FR, GB, GR, IE, IT, LU, MC, NL, PT, SE

OA : BF, BJ, CF, CG, CI, CM, GA, GN, GW, ML, MR, NE, SN, TD, TG

National : AL, AM, AT, AU, AZ, BA, BB, BG, BR, BY, CA, CH, CN, CU, CZ, DE, DK, EE, ES, FI, GB, GD, GE, GH, GM, HR, HU, ID, IL, IN, IS, JP, KE, KG, KP, KR, KZ, LC, LK, LR, LS, LT, LU, LV, MD, MG, MK, MN, MW, MX, NO, NZ, PL, PT, RO, RU, SD, SE, SG, SI, SK, SL, TJ, TM, TR, TT, UA, UG, US, UZ, VN, YU, ZW

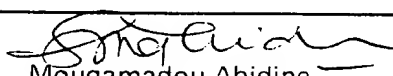
## ATTENTION

The applicant should carefully check the data appearing in this Notification. In case of any discrepancy between these data and the indications in the international application, the applicant should immediately inform the International Bureau.

In addition, the applicant's attention is drawn to the information contained in the Annex, relating to:

- ☒ time limits for entry into the national phase
- ☐ confirmation of precautionary designations
- ☒ requirements regarding priority documents

A copy of this Notification is being sent to the receiving Office and to the International Searching Authority.

The International Bureau of WIPO 34, chemin des Colombettes 1211 Geneva 20, Switzerland	Authorized officer:  Mougamadou Abidine
Facsimile No. (41-22) 740.14.35	Telephone No. (41-22) 338.83.38

## INFORMATION ON TIME LIMITS FOR ENTERING THE NATIONAL PHASE

The applicant is reminded that the "national phase" must be entered before each of the designated Offices indicated in the Notification of Receipt of Record Copy (Form PCT/IB/301) by paying national fees and furnishing translations, as prescribed by the applicable national laws.

The time limit for performing these procedural acts is **20 MONTHS** from the priority date or, for those designated States which the applicant elects in a demand for international preliminary examination or in a later election, **30 MONTHS** from the priority date, provided that the election is made before the expiration of 19 months from the priority date. Some designated (or elected) Offices have fixed time limits which expire even later than 20 or 30 months from the priority date. In other Offices an extension of time or grace period, in some cases upon payment of an additional fee, is available.

In addition to these procedural acts, the applicant may also have to comply with other special requirements applicable in certain Offices. **It is the applicant's responsibility** to ensure that the necessary steps to enter the national phase are taken in a timely fashion. Most designated Offices do not issue reminders to applicants in connection with the entry into the national phase.

For detailed information about the procedural acts to be performed to enter the national phase before each designated Office, the applicable time limits and possible extensions of time or grace periods, and any other requirements, see the relevant Chapters of Volume II of the PCT Applicant's Guide. Information about the requirements for filing a demand for international preliminary examination is set out in Chapter IX of Volume I of the PCT Applicant's Guide.

GR and ES became bound by PCT Chapter II on 7 September 1996 and 6 September 1997, respectively, and may, therefore, be elected in a demand or a later election filed on or after 7 September 1996 and 6 September 1997, respectively, regardless of the filing date of the international application. (See second paragraph above.)

Note that only an applicant who is a national or resident of a PCT Contracting State which is bound by Chapter II has the right to file a demand for international preliminary examination.

## CONFIRMATION OF PRECAUTIONARY DESIGNATIONS

This notification lists only specific designations made under Rule 4.9(a) in the request. It is important to check that these designations are correct. Errors in designations can be corrected where precautionary designations have been made under Rule 4.9(b). The applicant is hereby reminded that any precautionary designations may be confirmed according to Rule 4.9(c) before the expiration of 15 months from the priority date. If it is not confirmed, it will automatically be regarded as withdrawn by the applicant. There will be no reminder and no invitation. Confirmation of a designation consists of the filing of a notice specifying the designated State concerned (with an indication of the kind of protection or treatment desired) and the payment of the designation and confirmation fees. Confirmation must reach the receiving Office within the 15-month time limit.

## REQUIREMENTS REGARDING PRIORITY DOCUMENTS

~~For applicants who have not yet complied with the requirements regarding priority documents, the following is recalled~~

Where the priority of an earlier national, regional or international application is claimed, the applicant must submit a copy of the said earlier application, certified by the authority with which it was filed ("the priority document") to the receiving Office (which will transmit it to the International Bureau) or directly to the International Bureau, before the expiration of 16 months from the priority date, provided that any such priority document may still be submitted to the International Bureau before that date of international publication of the international application, in which case that document will be considered to have been received by the International Bureau on the last day of the 16-month time limit (Rule 17.1(a)).

Where the priority document is issued by the receiving Office, the applicant may, instead of submitting the priority document, request the receiving Office to prepare and transmit the priority document to the International Bureau. Such request must be made before the expiration of the 16-month time limit and may be subjected by the receiving Office to the payment of a fee (Rule 17.1(b)).

If the priority document concerned is not submitted to the International Bureau or if the request to the receiving Office to prepare and transmit the priority document has not been made (and the corresponding fee, if any, paid) within the applicable time limit indicated under the preceding paragraphs, any designated State may disregard the priority claim, provided that no designated Office may disregard the priority claim concerned before giving the applicant an opportunity to furnish the priority document within a time limit which is reasonable under the circumstances.

Where several priorities are claimed, the priority date to be considered for the purposes of computing the 16-month time limit is the filing date of the earliest application whose priority is claimed.

PCT

From the INTERNATIONAL BUREAU

NOTIFICATION CONCERNING  
SUBMISSION OR TRANSMITTAL  
OF PRIORITY DOCUMENT

(PCT Administrative Instructions, Section 411)

To:

TURNER, James, Arthur  
D Young & Co  
21 New Fetter Lane  
London EC4A 1DA  
ROYAUME-UNI

Date of mailing (day/month/year) 04 February 1999 (04.02.99)	<b>IMPORTANT NOTIFICATION</b> <i>CONFIRMED</i>
Applicant's or agent's file reference P/3952.WO	
International application No. PCT/GB98/03636	International filing date (day/month/year) 07 December 1998 (07.12.98)
International publication date (day/month/year) Not yet published	Priority date (day/month/year) 05 December 1997 (05.12.97)
Applicant IMPERIAL COLLEGE OF SCIENCE, TECHNOLOGY AND MEDICINE et al	

1. The applicant is hereby notified of the date of receipt (except where the letters "NR" appear in the right-hand column) by the International Bureau of the priority document(s) relating to the earlier application(s) indicated below. Unless otherwise indicated by an asterisk appearing next to a date of receipt, or by the letters "NR", in the right-hand column, the priority document concerned was submitted or transmitted to the International Bureau in compliance with Rule 17.1(a) or (b).
2. This updates and replaces any previously issued notification concerning submission or transmittal of priority documents.
3. An asterisk(\*) appearing next to a date of receipt, in the right-hand column, denotes a priority document submitted or transmitted to the International Bureau but not in compliance with Rule 17.1(a) or (b). In such a case, **the attention of the applicant is directed** to Rule 17.1(c) which provides that no designated Office may disregard the priority claim concerned before giving the applicant an opportunity, upon entry into the national phase, to furnish the priority document within a time limit which is reasonable under the circumstances.
4. The letters "NR" appearing in the right-hand column denote a priority document which was not received by the International Bureau or which the applicant did not request the receiving Office to prepare and transmit to the International Bureau, ~~and the applicant is directed to Rule 17.1(c) which provides that no designated Office may disregard the priority claim concerned before giving the applicant an opportunity, upon entry into the national phase, to furnish the priority document within a time limit which is reasonable under the circumstances.~~ **the attention of the applicant is directed to Rule 17.1(c) which provides that no designated Office may disregard the priority claim concerned before giving the applicant an opportunity, upon entry into the national phase, to furnish the priority document within a time limit which is reasonable under the circumstances.**

<u>Priority date</u>	<u>Priority application No.</u>	<u>Country or regional Office or PCT receiving Office</u>	<u>Date of receipt of priority document</u>
05 Dec 1997 (05.12.97)	9725878.4	GB	01 Febr 1999 (01.02.99)

The International Bureau of WIPO  
34, chemin des Colombettes  
1211 Geneva 20, Switzerland

Facsimile No. (41-22) 740.14.35

Authorized officer

Marc Salzman

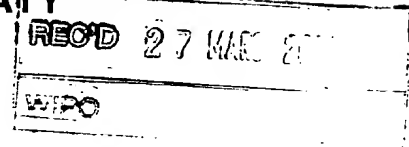
Telephone No. (41-22) 338.83.38

## PATENT COOPERATION TREATY

## PCT

## INTERNATIONAL PRELIMINARY EXAMINATION REPORT

(PCT Article 36 and Rule 70)





Applicant's or agent's file reference W003952	<b>FOR FURTHER ACTION</b> See Notification of Transmittal of International Preliminary Examination Report (Form PCT/IPEA/416)	
International application No. PCT/GB98/03636	International filing date (day/month/year) 07/12/1998	Priority date (day/month/year) 05/12/1997
International Patent Classification (IPC) or national classification and IPC C23C16/44		
Applicant IMPERIAL COLLEGE OF SCIENCE, TECHNOLOGY ... et al.		

1. This international preliminary examination report has been prepared by this International Preliminary Examining Authority and is transmitted to the applicant according to Article 36.
2. This REPORT consists of a total of 6 sheets, including this cover sheet.
- ☒ This report is also accompanied by ANNEXES, i.e. sheets of the description, claims and/or drawings which have been amended and are the basis for this report and/or sheets containing rectifications made before this Authority (see Rule 70.16 and Section 607 of the Administrative Instructions under the PCT).
- These annexes consist of a total of 9 sheets.

3. This report contains indications relating to the following items:

- I ☒ Basis of the report
- II ☐ Priority
- III ☐ Non-establishment of opinion with regard to novelty, inventive step and industrial applicability
- IV ☐ Lack of unity of invention
- V ☒ Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement
- VI ☐ Certain documents cited
- VII ☐ Certain defects in the international application
- VIII ☒ Certain observations on the international application

Date of submission of the demand  23/06/1999	Date of completion of this report  23. 03. 00
Name and mailing address of the international preliminary examining authority:   European Patent Office D-80298 Munich Tel. +49 89 2399 - 0 Tx: 523656 epmu d Fax: +49 89 2399 - 4465	Authorized officer  Werner, H  Telephone No. +49 89 2399 8571  

# INTERNATIONAL PRELIMINARY EXAMINATION REPORT

International application No. PCT/GB98/03636

## I. Basis of the report

1. This report has been drawn on the basis of *(substitute sheets which have been furnished to the receiving Office in response to an invitation under Article 14 are referred to in this report as "originally filed" and are not annexed to the report since they do not contain amendments.)*:

### Description, pages:

1-5 as received on 14/02/2000 with letter of 14/02/2000

### Claims, No.:

1-28 as received on 14/02/2000 with letter of 14/02/2000

### Drawings, sheets:

1/1 as originally filed

2. The amendments have resulted in the cancellation of:

- ☐ the description, pages:
- ☐ the claims, Nos.:
- ☐ the drawings, sheets:

3. ☒ This report has been established as if (some of) the amendments had not been made, since they have been considered to go beyond the disclosure as filed (Rule 70.2(c)):

**see separate sheet**

4. Additional observations, if necessary:

**INTERNATIONAL PRELIMINARY  
EXAMINATION REPORT**

International application No. PCT/GB98/03636

**V. Reasoned statement under Article 35(2) with regard to novelty, inventive step or industrial applicability; citations and explanations supporting such statement**

**1. Statement**

Novelty (N)	Yes:	Claims	4, 8, 10-12, 14, 19, 21-25
	No:	Claims	1, 5-6, 9, 13, 15-17, 26-28
Inventive step (IS)	Yes:	Claims	
	No:	Claims	1, 4-6, 8-17, 19, 21-28
Industrial applicability (IA)	Yes:	Claims	1, 4-6, 8-17, 19, 21-28
	No:	Claims	

**2. Citations and explanations**

**see separate sheet**

**VIII. Certain observations on the international application**

The following observations on the clarity of the claims, description, and drawings or on the question whether the claims are fully supported by the description, are made:

**see separate sheet**



## 1. SECTION I.3.

1.1. The amended claims 2, 3 (3 because it is dependent on claim 2), 7, 17, 18, and 20 as well as the description (page 2, line 5) filed with the letter dated 14.2.00, introduce subject-matter which extends beyond the content of the application as filed, contrary to Article 34(2)(b) PCT. The amendments concerned are the following:

- 1) "...an annular flow ... **about** the stream of droplets..." (in claims 2 and 7) and "...annular flow of..." (in claims 18 and 20). In the original application on page 2, lines 11-14 and Fig. 1 a **coaxial** nozzle assembly is disclosed. However, no *annular flow* has been disclosed in the original application.
- 2) "(the burner is) provided by the nozzle assembly" and "a fuel supply for supplying fuel to the nozzle assembly" (in claim 18). These features have not been disclosed in the original application.

Claims 2, 3, 7, 18, and 20 hence represent generalisations for which there is no basis in the original application. Therefore these claims are not considered regarding novelty, inventive step, and industrial applicability.

- 3) "a substrate holder for holding a substrate" (in claim 17 and page 2, line 5).

This feature has not been disclosed in the original application.

For the investigation regarding novelty, inventive step, and industrial applicability concerning claim 17 this feature was not considered.

## 2. SECTION V

2.1. Objections against Novelty (Art. 33(2) PCT):

2.1.1. Claims 1, 5, 6, 9, 13, 17, and 26 are not new according to Art. 33(2) PCT.

Document JP 56 005337 A discloses a method and an apparatus for depositing glass ( $\text{SiO}_2$ ) on a substrate comprising atomizing  $\text{Si}(\text{OC}_2\text{H}_5)_4$  to obtain a stream directed towards the substrate. An electrical field is applied between the nozzle and the target.  $\text{Si}(\text{OC}_2\text{H}_5)_4$  is mixed with hydrogen and the hydrogen is burned causing a flame directed towards the substrate and hence  $\text{Si}(\text{OC}_2\text{H}_5)_4$  is within the flame and thus being thermally decomposed before reaching the substrate. The  $\text{Si}(\text{OC}_2\text{H}_5)_4$  stream is divergent. A flow of an inert gas is delivered from the

nozzle towards the substrate. The substrate can be moved during the process of deposition.

2.1.2. Furthermore, also claims 15, 16, 27, and 28 are not new according to Art. 33(2) PCT since it is clear that the decomposition of  $\text{Si}(\text{OC}_2\text{H}_5)_4$  occurs in the flame, and hence away from or in the vicinity of the substrate.

2.2. Objections against inventive step (Art. 33(3) PCT):

2.2.1. Dependent claim 10, disclosing a multicomponent material, does not contain any features which, in combination with the features of any claim to which it refers, meet the requirements of the PCT in respect of inventive step (Art. 33(3) PCT), the reasons being as follows: Although document JP 56 005337 A does not disclose multicomponent materials, such materials are known to the person skilled in the art working in the area of semiconductors, and hence this feature is obvious.

2.2.2. Also the dependent claims 11 and 12 do not contain any features which, in combination with the features of any claim to which they refer, meet the requirements of the PCT in respect of inventive step, the reasons being as follows: Heating the substrate and using a sol precursor solution are two of several straightforward possibilities from which the skilled person would select, in accordance with circumstances, without the exercise of inventive skill, in order to solve the problem posed, and hence these features are obvious according to Art. 33(3) PCT.

2.2.3. Dependent claim 23 does not contain any features which, in combination with the features of any claim to which it refers, meet the requirements of the PCT in respect of inventive step (Art. 33(3) PCT), the reasons being as follows: Using a mesh does not seem to be of importance for solving the technical problem posed.

2.2.4. The features of dependent claims 24 and 25 have already been employed for the same purpose in a similar apparatus, see document JP 02 055241 A. It would therefore be obvious to the person skilled in the art to apply these features with corresponding effect to an apparatus as disclosed in JP 56 005337 A, thereby arriving at an apparatus according to claims 24 and 25.

2.2.5. Document JP 02 055241 A discloses an apparatus from which the subject-matter of claim 19 differs in that it discloses a nozzle assembly including an outlet for a

precursor **liquid** whereas in JP 02 055241 A the glass raw material is not precisely defined. However, the person skilled in the art knows that for such an apparatus the glass raw material must be a liquid or in the form of a fine powder. Furthermore, the person skilled in the art knows that in the present case the behaviour of a fine powder resembles a liquid. The glass raw material is reacted (hydrolysed) in the flame and the outlets are coaxial. Hence, the subject-matter of claim 19 does not involve an inventive step and does not satisfy the criterion set forth in Article 33(3) PCT.

- 2.2.6. The subject-matter of claims 21-22 does not involve an inventive step and does not satisfy the criterion set forth in Article 33(3) PCT because from document JP 02 055241 A taken in combination with document JP 56 005337 A the use of an extra outlet for cold gas is obvious. The difference to claims 21-22 is that in them the cold gas flow is between the precursor and the fuel whereas the combination of JP 56 005337 A and JP 02 055241 would result in a cold gas flow between the precursor-fuel and the oxygen. However, the technical effect would be the same, namely a possibility to control the combustion by the rate of cold gas.
- 2.2.7. For the same reasons as given in 2.2.5. the subject-matter of claim 4 does not involve an inventive step and does not satisfy the criterion set forth in Article 33(3) PCT.
- 2.2.8. For the same reasons as given in 2.2.6. the subject-matter of claim 8 does not involve an inventive step and does not satisfy the criterion set forth in Article 33(3) PCT.

### **3. SECTION VIII**

#### **3.1. Clarity (Art. 6 PCT):**

Claim 28 is written as a method claim. However, this seems to be erroneous because it refers back to apparatus claims. The investigation of claim 28 regarding novelty and inventive step has been performed assuming the claim is an apparatus claim.

## PCT

## INTERNATIONAL SEARCH REPORT

(PCT Article 18 and Rules 43 and 44)

Applicant's or agent's file reference <b>P/3952.W0</b>	<b>FOR FURTHER ACTION</b> see Notification of Transmittal of International Search Report (Form PCT/ISA/220) as well as, where applicable, item 5 below.	
International application No. <b>PCT/GB 98/ 03636</b>	International filing date (day/month/year) <b>07/12/1998</b>	(Earliest) Priority Date (day/month/year) <b>05/12/1997</b>
Applicant <b>IMPERIAL COLLEGE OF SCIENCE, TECHNOLOGY AND MEDICI</b>		

This International Search Report has been prepared by this International Searching Authority and is transmitted to the applicant according to Article 18. A copy is being transmitted to the International Bureau.

This International Search Report consists of a total of 4 sheets.  
☒ It is also accompanied by a copy of each prior art document cited in this report.

## 1. Basis of the report

- a. With regard to the **language**, the international search was carried out on the basis of the international application in the language in which it was filed, unless otherwise indicated under this item.
- ☐ the international search was carried out on the basis of a translation of the international application furnished to this Authority (Rule 23.1(b)).
- b. With regard to any **nucleotide and/or amino acid sequence** disclosed in the international application, the international search was carried out on the basis of the sequence listing:
- ☐ contained in the international application in written form.
- ☐ filed together with the international application in computer readable form.
- ☐ furnished subsequently to this Authority in written form.
- ☐ furnished subsequently to this Authority in computer readable form.
- ☐ the statement that the subsequently furnished written sequence listing does not go beyond the disclosure in the international application as filed has been furnished.
- ☐ the statement that the information recorded in computer readable form is identical to the written sequence listing has been furnished.

2. ☐ **Certain claims were found unsearchable** (See Box I).

3. ☐ **Unity of invention is lacking** (see Box II).

4. With regard to the **title**,

- ☒ the text is approved as submitted by the applicant.
- ☐ the text has been established by this Authority to read as follows:

5. With regard to the **abstract**,

- ☐ the text is approved as submitted by the applicant.
- ☒ the text has been established, according to Rule 38.2(b), by this Authority as it appears in Box III. The applicant may, within one month from the date of mailing of this international search report, submit comments to this Authority.

6. The figure of the **drawings** to be published with the abstract is Figure No.

- ☒ as suggested by the applicant.
- ☐ because the applicant failed to suggest a figure.
- ☐ because this figure better characterizes the invention.
- 1  
☐ None of the figures.

# INTERNATIONAL SEARCH REPORT

International application No.

PCT/GB 98/03636

## Box III TEXT OF THE ABSTRACT (Continuation of item 5 of the first sheet)

The abstract has to be changed as follows:  
Line 5, after "substrate" insert "(50)";  
line 6, after "outlet" insert "(60)";  
line 7, after "substrate" insert "(50)".

A. CLASSIFICATION OF SUBJECT MATTER  
IPC 6 C23C16/44

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)  
IPC 6 C23C

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	PATENT ABSTRACTS OF JAPAN vol. 005, no. 054 (C-050), 15 April 1981 & JP 56 005337 A (HITACHI LTD;OTHERS: 01), 20 January 1981 see abstract	1,2,6,7, 9-12,16
Y	see abstract	3,4,14, 15 8
A		
X	--- PATENT ABSTRACTS OF JAPAN vol. 006, no. 143 (C-117), 3 August 1982 & JP 57 067038 A (NIPPON TELEGR & TELEPH CORP), 23 April 1982 see abstract --- -/--	1,2,6,9, 16

☒ Further documents are listed in the continuation of box C.

☒ Patent family members are listed in annex.

\* Special categories of cited documents :

- "A" document defining the general state of the art which is not considered to be of particular relevance  
"E" earlier document but published on or after the international filing date  
"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)  
"O" document referring to an oral disclosure, use, exhibition or other means  
"P" document published prior to the international filing date but later than the priority date claimed

- "T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention  
"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone  
"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art.  
"&" document member of the same patent family

Date of the actual completion of the international search

26 May 1999

Date of mailing of the international search report

02/06/1999

Name and mailing address of the ISA

European Patent Office, P.B. 5818 Patentlaan 2  
NL - 2280 HV Rijswijk  
Tel. (+31-70) 340-2040, Tx. 31 651 epo nl,  
Fax: (+31-70) 340-3016

Authorized officer

Ekhult, H

## C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

Category	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
Y	PATENT ABSTRACTS OF JAPAN vol. 098, no. 001, 30 January 1998 & JP 09 235131 A (SHOWA ELECTRIC WIRE & CABLE CO LTD; NIPPON TELEGR & AMP; TELEPH CORP), 9 September 1997 see abstract ---	3,4
Y	PATENT ABSTRACTS OF JAPAN vol. 014, no. 227 (C-0718), 15 May 1990 & JP 02 055241 A (HITACHI CABLE LTD), 23 February 1990 see abstract ---	14,15
A	WO 97 21848 A (BAI WEI ; CHOY KWANG LEONG (GB); IMPERIAL COLLEGE (GB)) 19 June 1997 cited in the application ---	5
A	PATENT ABSTRACTS OF JAPAN vol. 013, no. 262 (C-608), 16 June 1989 & JP 01 065040 A (SUMITOMO ELECTRIC IND LTD), 10 March 1989 see abstract -----	13

## INTERNATIONAL SEARCH REPORT

Information on patent family members

International Application No

PCT/GB 98/03636

Patent document cited in search report	Publication date	Patent family member(s)	Publication date
WO 9721848 A	19-06-1997	GB 2308132 A	18-06-1997
		AU 1182997 A	03-07-1997
		CA 2240625 A	19-06-1997
		EP 0870075 A	14-10-1998